

# Magnetoresistance of Granular Ferromagnets - Observation of a Magnetic Proximity Effect?

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We have observed a superparamagnetic to ferromagnetic transition in films of isolated Ni grains covered by non-magnetic overlayers. The magnetoresistance (MR) of the films was measured as a function of the overlayer thickness. Initially, the granular Ni films exhibited negative MR curves peaked at  $H=0$ . As different materials were deposited onto the grains hysteresis developed in the MR. This behavior is ascribed to an increase of the typical domain size due to magnetic coupling between grains. The strength of the inter-grain coupling is found to correlate with the magnetic susceptibility of the overlayer material. We discuss possible mechanisms for this coupling and suggest that the data may reflect the existence of a magnetic proximity effect (analogous to the well-known effect in superconductivity) in which a ferromagnetic moment is induced in the metallic non-magnetic medium.

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The proximity effect is a well known phenomenon in superconductivity and has drawn a lot of interest both from the fundamental and the practical points of view. In a high transmission superconductor-normal metal contact the superconductive wave-function varies smoothly across the interface causing a suppression of the pair amplitude in the superconductor and an enhancement of superconductivity on the normal side. An analogue in magnetism, in which the magnetization varies smoothly across a ferromagnet-non magnetic metal interface, has been considered theoretically [1–4]. Experimentally, the observation of a magnetic proximity effect is much more challenging. In the first place the coherence length of a typical ferromagnet, such as Ni or Fe, is of the order of a few atomic spacings (this should be compared with  $\xi_s$  of thousands of Å in a conventional superconductor) and the means to measure the proximity effect are not as straightforward as in the superconducting case. Furthermore, one has to be able to distinguish between a “real” proximity effect and other magnetic phenomena such as magnetostatic interactions. One experimental approach in the past has been to study the suppression of magnetization in thin ferromagnets deposited on a normal metal substrate [5–8]. The first few monolayers were found to be magnetic “dead layers” exhibiting no ferromagnetic signal. In a different type of experiment Moodera et-al [9] used spin-polarized tunneling measurements to show that a finite spin polarization of electrons persists in a Au film coupled to a Fe layer up to a thickness of a few tens of Å. A question arises as to whether the spin polarization detection is indeed indicative of a ferromagnetic interaction inside the Au [10].

In this paper we describe an experiment which is designed to probe a potential magnetic analogue to the Josephson-like proximity coupling between superconductors across a SNS weak link. We measure the magne-

toresistance (MR) of a granular magnetic film covered by various non-magnetic overlayers and study the impact of these layers on the magnetic coupling between the grains.

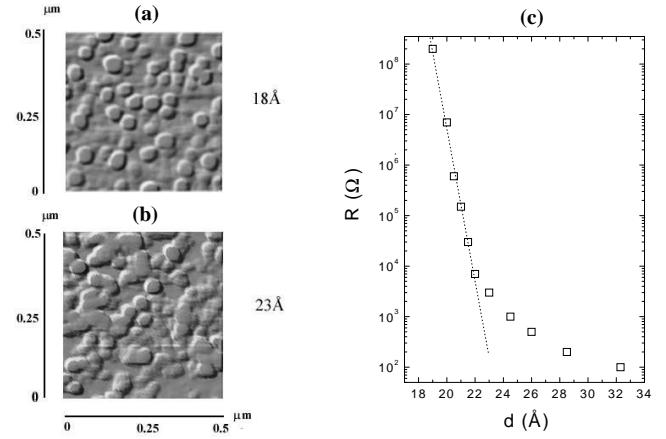


FIG. 1. Left: AFM micrographs of Ni films with nominal thickness of 18 Å and 23 Å evaporated on a room temperature Si-SiO substrate. Line scans show that the average height of the grains is 35 Å. Right: Sheet resistance versus nominal thickness of a quench-condensed Ni film.

The samples were prepared using the “quench-condensation” technique, i.e. evaporation on a cryo-cooled substrate under UHV conditions within the measurement apparatus. This method has a number of advantages for proximity effect experiments. First, the high vacuum in the system gives rise to barrier-free interfaces between different evaporated materials. These are essential for proximity effect observation. Furthermore, the low substrate temperature all but eliminates intermetallic diffusion, making the possibility of material alloying highly unlikely. Another advantage of this fab-

rication method is that it enables the evaporation of sequential layers of material *in-situ*; thus one can study the properties of a single sample as a function of the amount of deposited material while keeping the sample at low temperatures and in a UHV environment.

Our measurements were performed on thin layers of Ni quench-condensed onto a Si-SiO substrate. The morphology of such samples is illustrated in Figures 1a and 1b. For thin enough films the structure contains isolated grains with diameters of a few hundreds of Å [11] and heights of 30-40 Å. As more material is deposited, grains begin to coalesce with each other (Figure 1b). The average grain size thus increases and inter-grain spacing decreases until, beyond a percolation threshold, the film becomes continuous. This behavior is also demonstrated by the dependence of resistance on nominal film thickness (Figure 1c). For small thicknesses the resistance drops exponentially with thickness implying that the conduction mechanism is tunneling or hopping between grains. For larger thicknesses the film becomes continuous and the resistance crosses over to an ohmic  $1/d$  dependence.

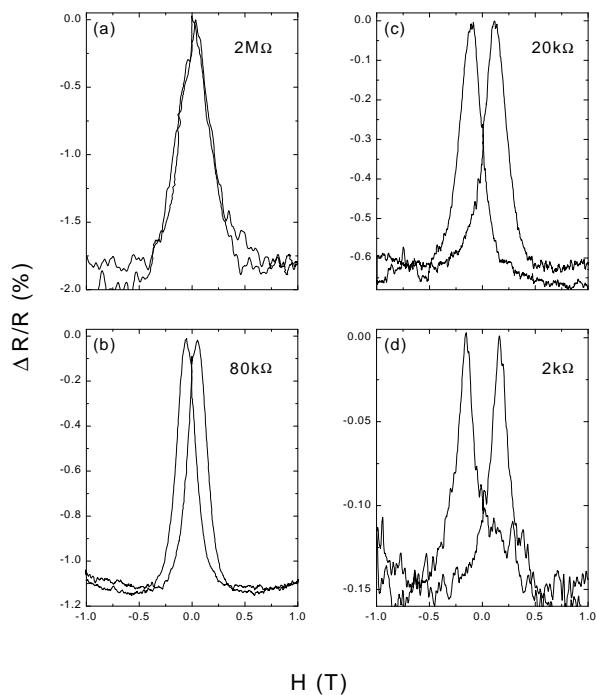


FIG. 2. Magnetoresistance curve at  $T=4.2\text{K}$  of a quench-condensed Ni film for different steps of the evaporation. The field was swept from  $-1\text{T}$  to  $1\text{T}$  and back. The nominal film thicknesses were  $20\text{\AA}$ (a),  $21\text{\AA}$ (b),  $21.8\text{\AA}$ (c) and  $25\text{\AA}$ (d).

Figure 2 shows MR curves of a Ni film for different deposition steps. The thinnest films exhibit curves which show a negative MR centered at  $H=0$  (Figure 2a). This

behavior is similar to that observed in other insulating granular ferromagnets prepared using different fabrication techniques [12–16] and is a result of spin dependent tunneling between grains which have randomly oriented magnetic moments [17]. Applying a field aligns these moments causing a resistance decrease. These thin films consist of small grains which are isolated from each other and are superparamagnetic at  $T=4\text{K}$ , thus, when the field is removed, the thermal energy is large enough to randomize the spin orientation and the MR curve does not exhibit hysteretic behavior.

As more material is deposited and the sheet resistance is reduced hysteresis develops in the curve, resulting in two resistance peaks at finite magnetic field. The position of the peaks shifts towards larger fields as the film thickens (figures 2b, 2c and 2d). Apparently, as the grains coalesce, the effective magnetic domain sizes become larger, the superparamagnetic blocking temperature rises and the film exhibits ferromagnetic behavior at  $T=4\text{K}$ . Indeed, the temperature dependence of these MR curves is consistent with well known relations for the superparamagnetic transition [18]. Adding material also causes the amplitude of the MR to decrease. This is a result of the percolation network for conductivity becoming denser, thus reducing the number of tunneling events which occur between grains with different oriented moments.

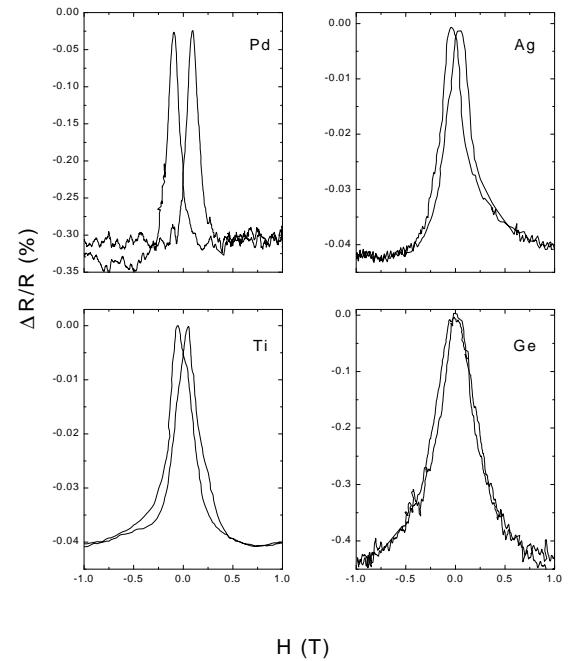


FIG. 3. MR curves at  $4.2\text{K}$  of  $20\text{\AA}$  thick films of granular Ni ( $R \approx 4\text{M}\Omega$ ) covered by different overlayers. The initial MR curve for the bare Ni was similar to that of figure 2a. For the Pd, Ti and Ag the overlayer thickness,  $d$ , is  $6\text{-}7\text{\AA}$  and the sheet resistance is  $2\text{k}\Omega$ . For Ge  $d = 65\text{\AA}$  and  $R = 10\text{K}\Omega$ .

Having observed the above effect of adding magnetic material to a granular magnetic film, we proceeded to study the effects of adding non-magnetic materials to superparamagnetic grains. We prepared films of isolated Ni grains (20 Å nominal thickness) which showed no hysteresis in the MR curve, and we added overlayers of Pd, Ti, Ag, or Ge *in-situ*. Figure 3 shows the MR curve of the samples in which an overlayer of 6 Å (or 65 Å in the Ge case) was added to the 20 Å Ni film. Despite the fact that Pd, Ti and Ag overlayers are non ferromagnetic, their presence gives rise to a hysteresis in the MR. Such a hysteresis is indicative of coupling between magnetic grains which were originally isolated. The coupling strength is different for the different overlayer materials.

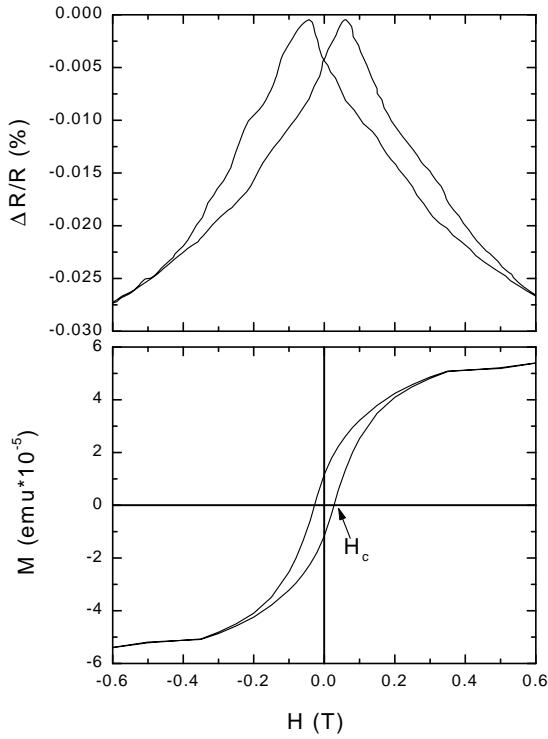


FIG. 4. MR and M-H curves taken at T=4.2K for a 23 Å thick Ni film evaporated at room temperature . The arrow marks the position of the coercive field,  $H_c$ .

We quantify the degree of hysteresis in the system by studying the coercive field,  $H_c$ , for which the magnetization in the system,  $M$ , equals zero. In a granular film this is the field required to totally randomize the magnetic orientations, hence it should correlate with the field at which the resistance peaks in the MR. Figure 4 compares the MR measurements and measurement of the magnetization versus field (M-H) hysteresis loop performed in a SQUID magnetometer. Though  $H_c$  differs slightly in these two experiments (the differences will be discussed in length elsewhere [18]) the behavior as a function of tem-

perature or film thickness is similar. The dependence of  $H_c$  on sheet resistance for the different overlaid materials, depicted in figure 5, clearly demonstrates that there is a correlation between the coupling strength of the medium and its magnetic susceptibility. Pd, which is a strong paramagnet ( $\chi \approx 11 \cdot 10^{-6} \text{emu/gr}\cdot\text{Oe}$ ) has an effect which is nearly as strong as that of adding Ni itself. Ti, a weaker paramagnet ( $\chi \approx 1.5 \cdot 10^{-6} \text{emu/gr}\cdot\text{Oe}$ ), has a significant but smaller coupling coefficient. Diamagnetic materials, such as Ag or Cu [19], also couple between the Ni grains, though the influence of diamagnetic overlayers is much weaker than that of paramagnets. In contrast, Ge, which is an insulator, does not induce any magnetic coupling at all. Notice that we have deposited a Ge overlayer which is much thicker than that of the metallic cases. Nevertheless, Ge seems to have no impact on the MR curve shape, in particular, it does not generate hysteretic behavior.

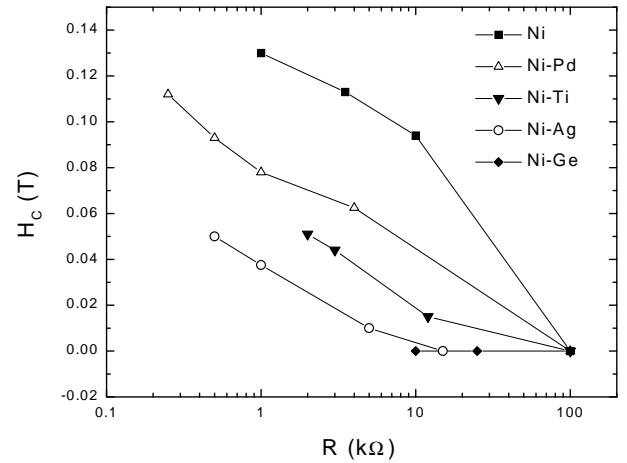


FIG. 5. Coercive field at T=4.2K as a function of sheet resistance for the different overlayer materials.

We have considered a number of possible scenarios for magnetic coupling through the normal medium. Classical magnetostatic coupling (dipole-dipole interaction between the grains) can be ruled out since the grains are isolated to begin with, hence, the magnetic interaction is obviously too weak to cause grain coupling. A strong candidate for the coupling mechanism is an exchange interaction between the grains mediated by the conduction electrons in the metal. Such behavior is seen in many magnetic heterostructures where magnetic layers are separated by non-magnetic spacers. In these systems it is found that the magnetic layers couple ferromagnetically or anti-ferromagnetically depending on the spacer thickness [20]. This behavior has been attributed to an RKKY-like interaction [21,22] which is oscillatory in space with a period of  $1/2k_f$ . There are a number of problems in trying to ascribe this mechanism to the re-

sults in our case. In the first place the RKKY process is very sensitive to the spacing between the magnets. Calculations show [22] that thickness roughness on the order of atomic spacing reduces the amplitude of short period oscillations by more than an order of magnitude. In our granular film there is no single grain spacing and the grains themselves are far from being atomically smooth, hence, the effect of an RKKY interaction is expected to average out. Furthermore, RKKY coupling is a Coulomb effect which depends mainly on the Fermi surface details and not on the magnetic properties of the spacing layer. There is no apparent reason why Ti should induce stronger coupling than Ag. In fact, Cu, which is the prototype spacer in multilayer systems, has hardly any effect in our granular samples. The fact that we observe a strong correlation of hysteresis with magnetic susceptibility implies that the relevant mechanism is one in which a magnetic moment is induced in the intermediate medium which, in turn, couples the magnetic grains. We note the similarity to superconductivity, where the proximity effect is enhanced for a normal material which is characterized by an internal strong electron-phonon coupling, such as a superconductor above its  $T_C$ .

In conclusion we have shown that isolated magnetic grains can be coupled via an intermediate medium which is not magnetic, resulting in a superparamagnetic-ferromagnetic crossover. This effect is stronger for paramagnetic media but exists also for diamagnets. We observe a clear relation between the coupling strength and the magnetic properties of the overlayer medium. This phenomenon bears a striking resemblance to an analogous experiment performed on granular superconductors in which an overlayer of Ag proximity couples isolated superconducting Pb grains [23]. The proximity coupling is expected to decay exponentially with the distance between grains. Within the framework of the current experiment we cannot yet provide an accurate evaluation of the relevant coherence length. Extraction of a coherence length will be pursued in future work. However, a rough estimation of the distance between our grains ( $\approx 10\text{\AA}$ ) points towards relatively short coherence lengths, especially in the diamagnetic layers. Hence, in high quality multilayers, the proximity effect can be expected to dominate for short scales (depending on the spacer magnetic properties) and the magnetic layers will couple ferromagnetically. For thicker spacers an RKKY-like exchange process, which decays like a power law, may set in and the sign of the coupling will oscillate with the thickness.

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